Theory of Incoherent Nuclear Inelastic Resonant Scattering of Synchrotron Radiation

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Abstract. The theory of incoherent nuclear resonant scattering of synchrotron radiation accompanied by absorption or emission of phonons in a crystal lattice is developed. The theory is based on the Maxwell's equations and time-dependent quantum mechanics under the condition of incoherent scattering of radiation by various nuclei. A concept of coherence in scattering processes, properties of the synchrotron radiation, and conditions of measurement are discussed. We show that employing the monochromator with a narrow bandwidth plays a decisive role. The equations for energy dependence and time evolution of scattered radiation are derived in detail for the case of the non-split nuclear levels. The effect of hyperfine interaction on the time spectra is also considered. We show that the Singwi and Sjölander theory of inelastic nuclear absorption, improved by the convolution with the instrumental function of the monochromator, is valid for the incoherent nuclear scattering in the case of integral over time measurements.

Key words: nuclear resonant scattering, inelastic scattering, phonons, coherence, energy spectrum, time dependence, monochromator.

1. Introduction

Nuclear resonant scattering of x rays attracts much interest after the Mössbauer's discovery of pure elastic nuclear resonant scattering [1]. The recoil energy is transferred to an entire crystal lattice, and the frequency of the scattered by nucleus radiation remains resonant. This gives an access to studies of coherent phenomena in nuclear scattering and stimulates a development of theory of coherent elastic nuclear resonant scattering (see the review of theoretical results in Refs. [2,3]. It became clear soon after the discovery that, because of the very small energy width of nuclear resonance, nuclear resonant scattering gives a good opportunity for studying dynamic of nuclear motions in crystals, especially thermal vibration and diffusion [4–6]. However, measurements of phonon spectra with radioactive sources were extremely difficult due to a small cross-section of inelastic processes.

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Recently the situation has drastically changed owing to the development of the synchrotron radiation sources of third generation such as ESRF (France), APS (USA) and SPring8 (Japan). A high brightness and spectral density of these sources, on the one hand, and a development of x-ray monochromator with energy bandwidth less than one millielectronvolt, on the other hand, give rise to numerous experimental studies of nuclear resonant inelastic absorption. These experiments deal with measurements of atomic fluorescence, which results from resonant absorption of incident photon by nucleus with simultaneous absorption or emission of phonons in solids. The review of experimental works and details of experimental methods are discussed in Ref. [7]. The analysis of the experimental results in the first works is based on the theory by Singwi and Sjölander [5], which is, strictly speaking, formulated only for the particular case of cubic Bravais crystal lattice. The general theory of nuclear inelastic absorption for anisotropic crystals is developed in Ref. [8].

We note that the scattering process is not discussed in these works. Only that part of nuclear absorption cross-section, which corresponds to absorption of photon by thermally vibrating nucleus is considered. The incident radiation is assumed as a monochromatic wave, i.e., the specific properties of the synchrotron radiation, the details of measurement and the time dependence of scattered radiation are not considered. One can believe that the theory is approximately valid for the nuclear resonant inelastic absorption accompanied by the atomic fluorescence, however, being not adapted to the real experimental conditions. As for the theory of nuclear resonant inelastic scattering (nuclear fluorescence) the problem remained open before recently. An attempt of developing the theory of both the nuclear fluorescence and the atomic fluorescence, which follow the process of inelastic nuclear excitation, is presented in Ref. [9] using methods of quantum electrodynamics and Green functions. However, in our opinion, the theory is not complete in the part where the details of experiment are discussed. Also the complicated technique does not allow obvious description of the scattering processes. The theory is difficult for understanding and requires special theoretical background.

This limitation can be overcome in frames of another approach which, after the Laue's [10] work, is widely used for description of various scattering processes. It is based on the Maxwell's equations and dielectic properties of medium. In many works (see, for example, Ref. [2]) nuclear resonant scattering is described by Maxwell's equation where a source of scattered radiation is treated as nuclear currents calculated by means of the time dependent perturbation theory of quantum mechanics [11]. In this work we present the theory of incoherent nuclear inelastic resonant scattering within this conventional approach. This allows us to take explicitly into account properties of the synchrotron radiation and conditions of measurements.

2. Maxwell's equation and coherence

For x rays with the energy about 10 keV and more, the electric $\mathbf{E}(\mathbf{r}, t)$ and magnetic $\mathbf{B}(\mathbf{r}, t)$ fields are strictly connected to each other, and the values of the modulo squared

field strength are identical after averaging over a period of oscillations . Therefore it is sufficient to consider an equation for any of these fields, or for some vector which allows one to determine both of them. In the quantum theory of radiation (see, for example, [12]) the vector potential $\mathbf{A}(\mathbf{r}, t)$ in the Coulomb gauge is used. It is chosen in such a way that

$$\nabla \cdot \mathbf{A} = 0 \,, \quad \mathbf{B} = \nabla \times \mathbf{A} \,, \quad \mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{A}}{\partial t} \,. \tag{1}$$

The Maxwell's equation for the vector potential can be written as

$$\nabla^2 \mathbf{A}(\mathbf{r}, t) - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{A}(\mathbf{r}, t) = -\frac{1}{c} \mathbf{j}(\mathbf{r}, t) \,, \tag{2}$$

where $\mathbf{j}(\mathbf{r}, t)$ is the current acting as a source of electromagnetic radiation, c is the speed of light.

2.1 Coherence as a possibility to observe interference.

The current $\mathbf{j}(\mathbf{r}, t)$ results from moving charged particles. If the particles move independently of the radiation, they are regarded as free sources of radiation. When the motion of particles is forced by incident radiation, we deal with re-radiation or scattering. In the latter case we deal with the induced current, which is proportional to the radiation field strength. The Maxwell's equation is linear relative to the field strength, and the induced currents for the x rays are also linear with high accuracy. Therefore the equation obeys the superposition principle for any ensemble of sources and scatterers. In other words, any fields and any scattering processes are coherent to each other.

This well known fact of classical optics should be stressed here, because sometimes one speaks about a principal incoherence of some sources or scattering processes. In reality, the problem of coherence absence is connected with conditions of measurement. Though the intensity of radiation from various sources or scatterers is always positive and must be only added, the interference term in the modulo squared total field strength has alternating signs in various time-space regions. As a rule, it strongly oscillates in time and space and its average value vanishes. Therefore, if experimental conditions allows only a measurement of average value of the interference term, the results cannot show coherent phenomena. However, if an experimental technique and properties of incident radiation allows one to observe the interference term, the processes are coherent. Thus, in order to observe the coherence one needs specific experimental conditions with essentially high spatial or temporal resolution of the detector.

For example, consider excitation of nuclei by x rays with extremely broad energy range containing all nuclear resonances. In this case, nuclear resonant scattering via various nuclear levels creates waves with so large difference in frequencies, that it would be impossible to record the interference term even with the best modern technique. Therefore, these acts of scattering should be considered as incoherent

[9]. However, scattering via various sub-levels of hyperfine structure of a single nuclear level leads to quantum beats in the time spectra with the period of about tens nanoseconds. Time resolution of available detectors allows measurements of resulting interference (see the review of experimental results in Ref. [13]). Thus, these quantum beats is a result of coherent phenomena.

Similar situations take place in space spectra. For example, if a source is strongly localized and a detector has sufficiently high spatial resolution, one can expect to record the interference phenomena even with x rays of high energy [14]. In general, the concept of coherence can be formulated as a possibility to observe the interference term in the intensity of radiation recorded from many sources and/or scatterers.

There exist particular conditions, when electric fields, resulted from scattering of primary radiation by very many nuclei, are added with the identical phase factor. Then the total intensity of scattered wave is proportional to N^2 where N is a number of nuclei. In this case the interference term can significantly exceed the average intensity, and the strength of the scattered wave can be comparable with the strength of the incident wave. The effective wave field, which excites the nuclei, is not known a priory in this case, and it should be calculated self-consistently. These conditions are called dynamical or multiple scattering. A conventional approach of dynamical theory of scattering supposes a calculation of the field inside the volume of scattering medium solving the self-consistent equations. The medium is considered as infinite or limited in one direction (lamina-like medium). The fields at both sides of the entrance surface of the sample must be identical (boundary conditions). The field at the exit surface is considered usually as that measured by a detector, although successive propagation of the wave in air may be important in some cases. In the dynamical theory, it is sometimes convenient to introduce a continuous spatial distribution of the scatterers and the medium polarization similarly to classical optics. A particular case of the dynamical theory is the kinematical theory, where the scattered wave remains much weaker than the incident wave, but scattering is still spatially coherent and depends on the shape of the sample.

Kinematical scattering is often identified with the first Born approximation although this is not completely correct. In the first Born approximation the boundary problem is not considered at all. The scatterers are assumed to be far away from both the source and the detector, whereas the scattering amplitudes are expected to be very small. If the experimental conditions allow one to record the interference term, the scattering process is coherent. If the interference term can be expected as negligible, it is sufficient to calculate the intensity of scattered wave from one nucleus. A total intensity of radiation becomes proportional to a number of nuclei in a target. In this approach, however, the act of scattering should not necessarily be connected to a single nucleus. In general, the incident wave illuminates many nuclei. The problem of localization or de-localization of excitation is, in fact, identical to the problem of phasing or de-phasing of the interference term in the total intensity of the radiation scattered by many nuclei.

In this work, we consider nuclear resonant scattering accompanied by absorption

or emission of phonons in a target. The process has relatively small cross-section. Therefore scattering events are collected within a large solid angle and are accumulated after many flashes of incident synchrotron radiation. These conditions do not allow us to record the interference between various nuclei. Thus, we can restrict ourselves by an analysis of the incoherent scattering. We will use the first Born approximation in the problem of scattering by a single nucleus. A summation over many nuclei leads to a trivial factor of a number of nuclei in the unit volume of the target. The task is solved in three independent steps. Firstly, we declare the properties of the incident radiation $\mathbf{A}_i(\mathbf{r}, t) = \mathbf{e}_i A_i(\mathbf{r}, t)$. Secondly, we calculate the current $\mathbf{j}_{fi}(\mathbf{r}, t)$, of nuclear resonant transitions. Finally, we calculate the scattered radiation $\mathbf{A}_f(\mathbf{r}, t) = \mathbf{e}_f A_f(\mathbf{r}, t)$, at large distance from the nucleus. Here $\mathbf{e}_i$ $\mathbf{e}_f$ are the unit vectors of polarizations.

2.2 Temporal and spatial structure of incident wave.

Properties of synchrotron undulator radiation are well known. They are complicated by significant relativistic phenomena. Instead of using them straightforwardly, we consider a simplified model, where the current of the source is strictly localized in time and space.

$$\nabla^2 A_i(\mathbf{r}, t) - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} A_i(\mathbf{r}, t) = -\frac{1}{c} \delta(\mathbf{r} - \mathbf{r}_0) \delta(t - t_s) \,. \tag{3}$$

Here $\mathbf{r}_0$ is the source position, and t_s is the instant of flash. Although a bunch of electrons in a storage ring consists of many electrons, the radiation from various electrons can be considered as incoherent. Therefore each event of nuclear scattering is initiated by incident radiation from a single electron. The width of the synchrotron radiation pulse (e.g., about 100 ps at the ESRF, Grenoble) does not enter the problem.

The solution of eq. (3) is obtained as the Fourier integral

$$A_i(\mathbf{r}, t) = \int \frac{d\omega}{2\pi} \exp(-i\omega[t - t_s]) \frac{\exp(i\omega|\mathbf{r} - \mathbf{r}_0|/c)}{4\pi c|\mathbf{r} - \mathbf{r}_0|} \,, \tag{4}$$

where the particular solution for the monochromatic source is applied as a spherical wave. The integral in (4) can be calculated analytically as follows

$$A_i(\mathbf{r}, t) = \frac{\delta(t - t_s - |\mathbf{r} - \mathbf{r}_0|/c)}{4\pi c|\mathbf{r} - \mathbf{r}_0|} \,. \tag{5}$$

This result has a simple physical meaning. Instantaneous localized excitation spreads in all directions from the point $\mathbf{r}_0$ and is distributed over the surface of the sphere of radius $|\mathbf{r} - \mathbf{r}_0|$. At the instant t the radius is equal to the distance $|\mathbf{r} - \mathbf{r}_0| = c(t - t_s)$ which the light covers during the time interval $(t - t_s)$. The intensity of the radiation is proportional to $|\mathbf{r} - \mathbf{r}_0|^{-2}$. The total intensity over the sphere area remains constant.

In further calculations, the integral presentation (4) is more convenient. At large distances from the source $|\mathbf{r}_0| \gg |\mathbf{r}|$ we can use the approximation

$$A_i(\mathbf{r}, t) \approx \frac{1}{4\pi c|\mathbf{r}_0|} \int \frac{d\omega}{2\pi} \exp(i\mathbf{k}_0'\mathbf{r} - i\omega[t - t_s - t_0]) \,, \tag{6}$$

where $t_0 = |\mathbf{r}_0|/c$ is the flight time through the distance $|\mathbf{r}_0|$ from the source to the origin of the our coordinate system where the nucleus is located, $\mathbf{k}'_0 = (\omega/c)\mathbf{s}_0$, $\mathbf{s}_0 = -\mathbf{r}_0/|\mathbf{r}_0|$. We use the approximation $|\mathbf{r} - \mathbf{r}_0| \approx |\mathbf{r}_0| + \mathbf{s}_0\mathbf{r}$. The expression (6) describes the set of plane monochromatic waves with all possible frequencies.

In reality, an electron in the storage ring is not localized in space but moves with the near-light speed. Therefore, the synchrotron radiation is strongly anisotropic. It is close to the spherical wave only in the electron coordinate system [15, 16]. At the point far from the source and in the laboratory coordinate system, it can be approximated by the Gauss beam directed along the speed of motion. Within its central part, however, the radiation wave field is close to the spherical wave which approximately coincides with the plane wave. Thus, the postulated spatial localization is consistent with experimental conditions. In order to estimate the temporal localization of single electron radiation flash, we can consider an effective time width as given by frequency spectrum of synchrotron radiation under typical experimental conditions and the uncertainty principle. The estimated time width turns out to be negligibly small compared to the nuclear life time. Thus, eq.(4) approximately describes the incident synchrotron radiation under the typical experimental conditions.

In experiments on nuclear inelastic scattering the bandwidth of radiation is limited by a monochromator. Let the transmission amplitude of the monochromator be $P(\omega - \omega_0)$, where ω_0 is the frequency at the maximum of transmissivity through the monochromatoron, while the instrumental function $I_M(\omega) = |P(\omega)|^2$ is centered at zero argument. The frequency band, provided by the monochromator, is sufficiently small to neglect the frequency dependence of the wave vector $\mathbf{k}'_0$, because the considered values of the vector $|\mathbf{r}|$ are small and comparable with displacements due to the thermal motion of the nucleus. As a result, we arrive to the following approximation for the amplitude of the incident radiation

$$A_i(\mathbf{r}, t) \approx \frac{\exp(i\mathbf{k}_0\mathbf{r} - i\omega_0[t - t_s - t_0])}{4\pi c|\mathbf{r}_0|} \int \frac{d\omega}{2\pi} \exp(-i\omega[t - t_s - t_0])P(\omega). \qquad (7)$$

Hereafter $\mathbf{k}_0 = (\omega_0/c)\mathbf{s}_0$.

The frequency integral in eq.(7) describes time evolution of the incident radiation flash after passing through the monochromator. It is determined by the response function of the monochromator

$$\tilde{P}(t) = \int \frac{d\omega}{2\pi} \exp(-i\omega t)P(\omega). \qquad (8)$$

According to general physical principles, $P(\omega)$ as a function of complex variable ω can have the poles only in the bottom half-plane. Therefore the response function $\tilde{P}(t)$ vanishes for $t < 0$. In other words, it describes the retarded response. The nucleus perceives the radiation flush at the instant $t = t_s + t_0$, which is by t_0 later than t_s – the instant of the flash at the source . Time t_0 is required for light to travel the distance $|\mathbf{r}_0|$. The accurate time profile of the flash perceived by the nucleus is determined by the monochromator and is described by the function $\tilde{P}(t)$.

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2.3 Temporal and spatial structure of scattered wave.

It will be shown below that the current of nuclear transition can be expressed as an integral over a limited range of frequencies near the middle frequency ω_1,

$$\mathbf{e}_f \mathbf{j}_{fi}(\mathbf{r}, t) = \int \frac{d\omega}{2\pi} \exp[-i(\omega_1 + \omega)t] J(\mathbf{r}, \omega). \tag{9}$$

Therefore we can write the amplitude of the scattered wave at the detector position also as a spherical monochromatic waves:

$$A_f(\mathbf{r}_1, t_d) = \int \frac{d\omega}{2\pi} \exp[-i(\omega_1 + \omega)t_d] \int d\mathbf{r} \frac{\exp(ik_1'|\mathbf{r}_1 - \mathbf{r}|)}{4\pi c |\mathbf{r}_1 - \mathbf{r}|} J(\mathbf{r}, \omega), \tag{10}$$

where t_d is the instant of arriving the radiation flash to the detector, and $k_1' = (\omega_1 + \omega)/c$.

We assume that the distance $|\mathbf{r}_1|$ between the detector and the scatterer is much larger than the typical displacement of the nucleus $|\mathbf{r}|$ due to thermal vibrations. We place again the centre of the scatterer at the origin of the coordinate system and use the relation $\mathbf{r}_1 - \mathbf{r} = |\mathbf{r}_1|\mathbf{s}_1 - \mathbf{r}$, where $\mathbf{s}_1 = \mathbf{r}_1/|\mathbf{r}_1|$. Then, applying an approximation $|\mathbf{r}_1 - \mathbf{r}| \approx |\mathbf{r}_1| - \mathbf{s}_1\mathbf{r}$, we arrive to

$$A_f(\mathbf{r}_1, t_d) = \int \frac{d\omega}{2\pi} \frac{\exp[-i(\omega_1 + \omega)(t_d - t_1)]}{4\pi c |\mathbf{r}_1|} \int d\mathbf{r} \exp(-i\mathbf{k}_1\mathbf{r}) J(\mathbf{r}, \omega), \tag{11}$$

Here $\mathbf{k}_1$ is the scattering vector, $\mathbf{k}_1 = k_1 \mathbf{s}_1 = k_1 \mathbf{r}_1/|\mathbf{r}_1|$. Its modulus is $k_1 = \omega_1/c$, because we can neglect small frequency ω due to the same reason as for the incident radiation. The value $t_1 = |\mathbf{r}_1|/c$ is the time interval required for light to travel the distance $|\mathbf{r}_1|$ from the nucleus to the detector.

Thus, the scattered radiation from single nucleus is described by an anisotropic spherical wave, which shape is determined by properties of the nucleus.

3. Quantum current of nuclear transitions.

The scatterer is the nucleus located in the node of crystal lattice of the target. The state of scatterer can be described in terms of the collective state of the centres of mass of all nuclei in the crystal $|\chi_i\rangle$ and the internal states of the nucleus $|\phi_i\rangle$. Consider first the internal nuclear transitions.

3.1 The Schrödinger equation.

The current $\mathbf{j}(\mathbf{r}, t)$ is the quantum mechanical average value of the current density operator over the states in presence of the electromagnetic field of radiation. Since the notation $|\phi_j\rangle$ is used for the stationary states of the nucleus we will denote the wave function in the presence of the radiation field by $|\psi_j(t)\rangle$.

We consider a 3-level system with $|\phi_i\rangle f_i(t)$ as the initial state, $|\phi_j\rangle f_j(t)$ as the intermediate state and $|\phi_f\rangle f_f(t)$ as the final state of the nucleus. The equation for the quasi-stationary states is

$$\hat{H}_0|\phi_j\rangle = E_j|\phi_j\rangle\,, \quad f_j(t) = \exp(-i\omega_j t - \gamma_j t)\,, \quad \omega_j = E_j/\hbar\,, \quad \gamma_j = \Gamma_j/2\hbar\,. \quad (12)$$

Here $\hat{H}_0$ is the Hamiltonian of the nucleus. We explicitly take into account a decay of the excited states with time. For the sake of simplicity, in this section we assume, that the hyperfine splitting of the nuclear levels is absent. In the problem of the energy spectrum of inelastic nuclear scattering with participation of phonons the hyperfine splitting is not essential at all. It influences only the temporal characteristics of scattering. This question will be analyzed later.

The ground state of the nucleus $|\psi_i(t)\rangle$ is perturbed by the field of incident radiation $\mathbf{A}_i(\mathbf{r}, t)$. The Hamiltonian $\hat{H}_{int}(t)$ of interaction of the nucleus with the radiation field is small and can be taken into account in frames of perturbation theory. In general, the perturbed ground state contains small contributions from other nuclear states:

$$|\psi_i(t)\rangle = |\phi_i\rangle f_i(t) + a_{ji}(t)|\phi_j\rangle f_j(t) + \cdots\,. \quad (13)$$

This function is the solution of the Schrödinger equation

$$i\hbar\frac{\partial \psi_i(t)}{\partial t} = [\hat{H}_0 + \hat{H}_{int}(t)]\psi_i(t)\,. \quad (14)$$

A substitution of (13) into (14) leads to

$$i\hbar\frac{\partial f_i}{\partial t}\phi_i + a_{ji}i\hbar\frac{\partial f_j}{\partial t}\phi_j + i\hbar\frac{\partial a_{ji}}{\partial t}\phi_j f_j \approx \hat{H}_0\phi_i f_i + a_{ji}\hat{H}_0\phi_j f_j + \hat{H}_{int}(t)\phi_i f_i\,. \quad (15)$$

Since the first and second terms from both sides are equal to each other, we obtain

$$i\hbar\frac{\partial a_{ji}}{\partial t} = \langle\phi_j|\hat{H}_{int}(t)|\phi_i\rangle f_i(t)f_j^{-1}(t)\,. \quad (16)$$

Assuming the adiabatic inclusion of the interaction, i.e. $\hat{H}_{int}(-\infty) = 0$, we arrive to expression

$$a_{ji}(t) = \frac{1}{i\hbar}\int_{-\infty}^{t} dt' \exp(i\omega_{ji}t' + \gamma_j t')\langle\phi_j|\hat{H}_{int}(t')|\phi_i\rangle\,. \quad (17)$$

Here $\omega_{ji} = \omega_j - \omega_i$. We explicitly took into account that $\gamma_i = 0$.

3.2 Induced current as a source of scattered radiation.

Let us consider the matrix element of the current density operator for the transition of the second order from the initial state i into the state f assuming that the transition is realized through the intermediate state j which is excited by a radiation transition from i into j. The final state f must be considered with zero radiative width, because

a finite width describes transition probability and, therefore, has sense only for the intermediate state. Applying the Fourier transformation we have

$$\mathbf{j}_{fi}(\mathbf{r},t) = \int \frac{d\mathbf{k}}{(2\pi)^3} e^{i\mathbf{k}[\mathbf{r}-\mathbf{r}_n(t)]} e^{i\omega_f t} \langle \phi_f | \hat{\mathbf{j}}_n(\mathbf{k}) | \psi_i(t) \rangle \,, \tag{18}$$

where $\mathbf{r}_n(t)$ is the coordinate of the vibrated nucleus. Resonant scattering is described by the perturbed term in (13). In accord with (17) we have

$$\mathbf{j}_{fi}(\mathbf{r},t) = \frac{\exp(i\omega_{fi}t)}{i\hbar} \int \frac{d\mathbf{k}}{(2\pi)^3} \int_{-\infty}^{t} dt' e^{i\mathbf{k}[\mathbf{r}-\mathbf{r}_n(t)]} \langle \phi_f | \hat{\mathbf{j}}_n(\mathbf{k}) | \phi_j \rangle$$
$$\times \langle \phi_j | \hat{H}_{int}(t') | \phi_i \rangle \exp[i(-\omega_{ji} + i\gamma_j)(t-t')] \,. \tag{19}$$

The Hamiltonian of interaction is well known. It is convenient to write it in the form

$$\hat{H}_{int}(t') = -\frac{1}{c} \int d\mathbf{r}' \, \hat{\mathbf{j}}_n(\mathbf{r}'-\mathbf{r}_n) \mathbf{A}_i(\mathbf{r}',t')$$
$$= -\frac{1}{c} \int d\mathbf{r}' \int \frac{d\mathbf{k}'}{(2\pi)^3} e^{i\mathbf{k}'(\mathbf{r}'-\mathbf{r}_n(t'))} \mathbf{e}_i \hat{\mathbf{j}}_n(\mathbf{k}') A_i(\mathbf{r}',t') \,, \tag{20}$$

where, as above, $\mathbf{e}_i$ is the unit vector of polarization of incident radiation. Then, the projection of the current density on the unit vector of polarization of the scattered wave may be written in the form

$$\mathbf{e}_f \mathbf{j}_{fi}(\mathbf{r},t) = \int_{-\infty}^{t} dt' \int d\mathbf{r}' \, M_{fi}(\mathbf{r},t;\mathbf{r}',t') A_i(\mathbf{r}',t') \,, \tag{21}$$

where we introduce the scattering matrix

$$M_{fi}(\mathbf{r},t;\mathbf{r}',t') = \frac{i \exp(i\omega_{fi}^{(n)}t)}{\hbar c} \int \frac{d\mathbf{k}d\mathbf{k}'}{(2\pi)^6} e^{i\mathbf{k}\mathbf{r}+i\mathbf{k}'\mathbf{r}'} N_{fi}(\mathbf{k},\mathbf{k}')$$
$$\times \exp[i(-\omega_{ji}^{(n)} + i\gamma_j)(t-t')] e^{-i\mathbf{k}\mathbf{r}_n(t)} e^{-i\mathbf{k}'\mathbf{r}_n(t')} \,. \tag{22}$$

Hereafter the frequencies $\omega_{fi}^{(n)}$ related to the nuclear transitions are marked by the index (n) and we use notation

$$N_{fi}(\mathbf{k},\mathbf{k}') = \langle \phi_f | \mathbf{e}_f \hat{\mathbf{j}}_n(\mathbf{k}) | \phi_j \rangle \langle \phi_j | \mathbf{e}_i \hat{\mathbf{j}}_n(\mathbf{k}') | \phi_i \rangle \,. \tag{23}$$

Expression (22) for the scattering matrix contains the coordinates of the nucleus at the instant of excitation t' and at the instant of relaxation t.

The nuclear coordinates depend on the time owing to thermal vibrations. As known, the thermal vibrations of atoms in a crystal lattice exhibit distinct quantum behaviour, thus they must be described as absorption and emission of phonons. Therefore we will consider the coordinate $\mathbf{r}_n$ as the operator and introduce the wave function of phonons.

$$e^{-i\mathbf{k}\mathbf{r}_n(t)} e^{-i\mathbf{k}'\mathbf{r}_n(t')} = \sum_m \langle \chi_f | e^{-i\mathbf{k}\mathbf{r}_n} | \chi_m \rangle \langle \chi_m | e^{-i\mathbf{k}'\mathbf{r}_n} | \chi_i \rangle \exp(i\omega_{fm}^{(p)}t + i\omega_{mi}^{(p)}t')$$
$$= \exp(i\omega_{fi}^{(p)}t) L_{fi}(\mathbf{k},\mathbf{k}',t-t') \,. \tag{24}$$

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Here index (p) shows that the frequency is related to the phonon system. The sum over intermediate states of the phonon system is required in order to describe an evolution of the system from the instant t' into the instant t. The function L_{fi} depends only on the time interval and is defined by expression

$$L_{fi}(\mathbf{k}, \mathbf{k}', t) = \sum_m \langle \chi_f | e^{-i\mathbf{k}\mathbf{r}_n} | \chi_m \rangle \langle \chi_m | e^{-i\mathbf{k}'\mathbf{r}_n} | \chi_i \rangle \exp(-i\omega_{mi}^{(p)}t) \,. \tag{25}$$

As a result, the scattering matrix takes the form

$$M_{fi}(\mathbf{r}, t; \mathbf{r}', t') = \frac{i \exp(i\omega_{fi}t)}{\hbar c} \exp[i(-\omega_r + i\gamma)(t - t')]$$
$$\times \int \frac{d\mathbf{k}d\mathbf{k}'}{(2\pi)^6} e^{i\mathbf{k}\mathbf{r}+i\mathbf{k}'\mathbf{r}'} N_{fi}(\mathbf{k}, \mathbf{k}') L_{fi}(\mathbf{k}, \mathbf{k}', t - t') \,, \tag{26}$$

where we introduce the total transition frequency $\omega_{fi} = \omega_{fi}^{(n)} + \omega_{fi}^{(p)}$, the resonance frequency $\omega_r = \omega_{ji}^{(n)}$ and omit the index of the width of the excited state $\gamma = \gamma_j$. Expression (26) is close to eq.(25) of Ref. [9]. However, here it is derived for 3-level system within standard quantum mechanics approach. A summation over the intermediate states of the nucleus, as made in Ref. [9], does not have sense because the resonance can be realized only for one transition.

Substituting eqs. (26) and (7) into eq. (21) and making identical transformations we obtain the explicit form for the function $J(\mathbf{r}, \omega)$ in eq. (9) as follows

$$J(\mathbf{r}, \omega) = P(\omega)\frac{i \exp[i(\omega_0 + \omega)(t_s + t_0)]}{4\pi\hbar c^2|\mathbf{r}_0|} \int \frac{d\mathbf{k}}{(2\pi)^3} \exp(i\mathbf{k}\mathbf{r}) N_{fi}(\mathbf{k}, -\mathbf{k}_0)$$
$$\times \tilde{L}_{fi}(\mathbf{k}, -\mathbf{k}_0, \omega_0 + \omega - \omega_r + i\gamma) \tag{27}$$

Here $\omega_1 = \omega_0 - \omega_{fi}$ is the middle frequency of the scattered wave. We also introduced the function

$$\tilde{L}_{fi}(\mathbf{k}, \mathbf{k}', \omega) = \int_0^\infty dt \exp(i\omega t) L_{fi}(\mathbf{k}, \mathbf{k}', t) \,. \tag{28}$$

4. Properties of scattered radiation.

Substituting (27) in (11) we obtain the vector potential of the scattered wave as follows

$$A_f(\mathbf{r}_1, t_d) = \frac{\exp[-i\omega_1(t_d - t_1)] \exp[i\omega_0(t_s + t_0)]}{(4\pi)^2 c^3 \hbar |\mathbf{r}_0||\mathbf{r}_1|} N_{fi}(\mathbf{k}_1, -\mathbf{k}_0)$$
$$\times \int \frac{d\omega}{2\pi} \exp(-i\omega t) \tilde{L}_{fi}(\mathbf{k}_1, -\mathbf{k}_0, \omega_0 + \omega - \omega_r + i\gamma) P(\omega) \,. \tag{29}$$

Here $t = t_d - t_s - t_0 - t_1$ is the time delay of radiation in the monochromator and in the nucleus, because t_d is the time instant of arrival of the scattered flash to the detector, whereas $(t_s + t_0 + t_1)$ have sense of the time instant of arrival of the flash propagating in vacuum along the identical trajectory. The definition (28) states

that the function $\tilde{L}_{fi}(\mathbf{k}_1, -\mathbf{k}_0, \omega)$ has the poles only in the bottom half-plane of the complex variable ω. Therefore the strength of the scattered radiation as function of the delay time t vanishes for $t < 0$. As a result, we obtain general expression for intensity of scattered radiation as a function of the delay time t and the mean frequency ω_0 of the monochromator transmissivity as

$$I(\omega_0, t) = I_0 I_N \left| \int \frac{d\omega}{2\pi} \exp(-i\omega t) \tilde{L}_{fi}(\mathbf{k}_1, -\mathbf{k}_0, \omega_0 + \omega - \omega_r + i\gamma) P(\omega) \right|^2, \qquad (30)$$

where

$$I_0 = \frac{\omega_1^2}{(4\pi)^4 c^8 \hbar^2 r_0^2 r_1^2}, \qquad I_N = |N_{fi}(\mathbf{k}_1, -\mathbf{k}_0)|^2. \qquad (31)$$

In measurements of nuclear inelastic scattering, the signal is accumulated over many pulses of synchrotron radiation and resulted from scattering by many nuclei. Therefore the above expression must be summed over the final states (index f) and averaged over the initial states (index i) of the phonon system. We will denote such intensity as $\overline{I(\omega_0, t)}$ and call the average intensity.

4.1 Energy dependence of scattered radiation.

It is instructive to analyze first the complete energy spectrum of the scattered radiation when the measurement is performed during the total delay time interval from zero to infinity and the integral intensity is

$$I_{int}(\omega_0) = \int_0^\infty I(\omega_0, t) dt. \qquad (32)$$

Since $I(t) = 0$ at $t < 0$, the integration limits can be formally replaced by $(-\infty, \infty)$. Taking into account that expression (30) is the modulo squared of the Fourier integral over frequency, we use the Parseval's theorem and obtain straightforwardly

$$I_{int}(\omega_0) = I_0 I_N \int \frac{d\omega}{2\pi} \left| \tilde{L}_{fi}(\mathbf{k}_1, -\mathbf{k}_0, \omega_0 - \omega_r + \omega + i\gamma) \right|^2 I_M(\omega). \qquad (33)$$

Thus, the time-integrated intensity of scattered radiation as a function of the middle frequency ω_0 of incident radiation is a convolution of the energy spectrum of inelastic scattering for the case of monochromatic incident wave with the instrumental function as the energy spectrum of the monochromator $I_M(\omega) = |P(\omega)|^2$.

In order to calculate the averaged intensity $\overline{I_{int}(\omega_0)}$ we have to average the function $\left| \tilde{L}_{fi}(\mathbf{k}_1, -\mathbf{k}_0, \omega + i\gamma) \right|^2$. Substituting (25) in (28), we obtain

$$\tilde{L}_{fi}(\mathbf{k}_1, -\mathbf{k}_0, \omega + i\gamma) = -\sum_m \frac{\langle \chi_f | e^{-i\mathbf{k}_1 \mathbf{r}_n} | \chi_m \rangle \langle \chi_m | e^{i\mathbf{k}_0 \mathbf{r}_n} | \chi_i \rangle}{(\omega - \omega_{mi}^{(p)} + i\gamma)}. \qquad (34)$$

The square modulus of the right-hand side must be summarized over the final states that leads to the expression

$$\sum_{f,m,m'} \frac{\langle \chi_f | e^{-i\mathbf{k}_1 \mathbf{r}_n} | \chi_m \rangle \langle \chi_m | e^{i\mathbf{k}_0 \mathbf{r}_n} | \chi_i \rangle}{(\omega - \omega_{mi}^{(p)} + i\gamma)} \frac{\langle \chi_i | e^{-i\mathbf{k}_0 \mathbf{r}_n} | \chi_{m'} \rangle \langle \chi_{m'} | e^{i\mathbf{k}_1 \mathbf{r}_n} | \chi_f \rangle}{(\omega - \omega_{m'i}^{(p)} - i\gamma)}. \qquad (35)$$

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Here the sum over f can be calculated independently

$$\sum_f \langle \chi_{m'} | e^{i\mathbf{k}_1\mathbf{r}_n} | \chi_f \rangle \langle \chi_f | e^{-i\mathbf{k}_1\mathbf{r}_n} | \chi_m \rangle = \langle \chi_{m'} | | \chi_m \rangle = \delta_{mm'} . \tag{36}$$

Then the dependence on the direction of the scattered wave vanishes and, instead of the triple sum, we obtain only a sum over m .

Replacing index $m \to f$, we may present the averaged intensity in the form

$$\overline{I_{int}(\omega_0)} = I_0 I_N \int \frac{d\omega}{2\pi} I_{SS}(\omega_0 - \omega_r + \omega) \, I_M(\omega) , \tag{37}$$

where

$$I_{SS}(\omega) = \langle \sum_f \left| \tilde{L}_{fi}(\mathbf{k}_1, -\mathbf{k}_0, \omega + i\gamma) \right|^2 \rangle_T = \sum_{i,f} \frac{g_i \left| \langle \chi_f | \exp(i\mathbf{k}_0\mathbf{r}_n) | \chi_i \rangle \right|^2}{(\omega_{fi}^{(p)} - \omega)^2 + \gamma^2} . \tag{38}$$

Here $\langle \cdots \rangle_T$ means the thermal averaging over the initial states, and g_i is the weight of i-th initial state of the phonon system.

Equation (37) is a standard recipe of taking into account the instrumental function (see, for example, Refs. [8, 18]). Eq. (38) for the integral intensity of *scattered* radiation coincides with the known expression for cross-section of nuclear resonant *absorption*, used by Singwi and Sjölander (see eq.(1) in Ref. [5]). An important property of this formula is that absorption or emission of phonons takes place simultaneously with resonant absorption of incident photon by nucleus, i.e., at the instant of nuclear excitation. The intensity of scattered radiation depends only on the wave vector $\mathbf{k}_0$, whereas the wave vector $\mathbf{k}_1$ does not influence the energy spectrum. As follows from eq.(36), this feature is a logical sequence of the procedure of summation over all the possible final states of the phonon system. This treatment corresponds to measurement of scattered quanta independently of their energy and directions of propagation. If an experimental setup would contain additional elements allowing selection of scattered radiation with a particular energy, or if scattered radiation would be accepted within essentially small solid angle, we have to use instead of eq.(29) some more complicated expressions with an explicit integration over frequency ω_1. Besides that, we would have to use the theory of coherent scattering, i.e., to take into account interference between various nuclei. In general, energy dependence of radiation scattered into some particular directions may then differ from the averaged value.

Thus, under the conditions of entire averaging over angle and energy of scattered radiation, the energy dependence of scattered radiation coincide entirely with the energy dependence of nuclear inelastic absorption. This conclusion agrees with the experimental results of Ref. [17]. One of the reason of this coincidence is that spontaneous emission of photon during de-excitation of a single nucleus does not depend on the resonance condition of preceding nuclear excitation. In particular, it does not depend on the phonon state, which has determined conditions of nuclear excitation. That is why, the energy spectrum of incoherent nuclear inelastic scattering

does not differ from those of nuclear inelastic absorption. At the first step, a nucleus is excited by an incident quantum with some energy transfer to absorption or emission of phonons. The following evolution of excited nucleus may vary, but it does not influence the dependence of recorded radiation on the energy of incident radiation.

Following Ref. [5], it is convenient to represent the function $I_{SS}(\omega)$ as integral over time. Below we use slightly different, than in Ref. [5], way of derivation which is more useful for us. Let us write the expression (38) as follows

$$I_{SS}(\omega) = \sum_{i,f} g_i \, |\langle \chi_f | \, \exp(i\mathbf{k}_0\mathbf{r}_n) | \chi_i \rangle|^2 \tilde{R}(\omega_{fi}^{(p)} - \omega) \,, \tag{39}$$

where

$$\tilde{R}(\omega) = \frac{1}{(\omega^2 + \gamma^2)} = \int dt \exp(i\omega t) \, R(t) \,. \tag{40}$$

Substituting the Fourier integral (40) in eq.(39) we obtain

$$I_{SS}(\omega) = \int dt \exp(-i\omega t) \, F(\mathbf{k}_0, t) \, R(t) \,, \tag{41}$$

where the time correlation function $F(\mathbf{k}_0, t)$ depends on the lattice vibrations and is determined by

$$\begin{aligned} F(\mathbf{k}_0, t) &= \sum_i g_i \sum_f \langle \chi_i | \, \exp(-i\mathbf{k}_0\mathbf{r}_n) | \chi_f \rangle \langle \chi_f | \, \exp(i\mathbf{k}_0\mathbf{r}_n) | \chi_i \rangle \exp(i\omega_{fi}^{(p)} t) \\ &= \sum_i g_i \langle \chi_i | \, \exp(-i\mathbf{k}_0\mathbf{r}_n(0)) \, \exp(i\mathbf{k}_0\mathbf{r}_n(t)) | \chi_i \rangle \\ &= \langle \exp(-i\mathbf{k}_0\mathbf{r}_n(0)) \, \exp(i\mathbf{k}_0\mathbf{r}_n(t)) \rangle_T \,. \end{aligned} \tag{42}$$

The function $R(t)$ may be calculated in explicit form

$$R(t) = \frac{\exp(-\gamma|t|)}{2\gamma} \,. \tag{43}$$

As a result, we arrive to expression

$$\overline{I_{int}(\omega_0)} = I_0 I_N \int dt \exp(-i[\omega_0 - \omega_r]t) F(\mathbf{k}_0, t) \, R(t) \, \tilde{I}_M(t) \,, \tag{44}$$

where

$$\tilde{I}_M(t) = \int \frac{d\omega}{2\pi} \exp(-i\omega t) I_M(\omega) \,. \tag{45}$$

In Refs. [8, 18] similar expression was used in order to obtain phonon density of states from energy dependence of nuclear inelastic absorption. Our results show that, if incoherent nuclear inelastic scattering is ideally integrated over time, its energy dependence is identical to that of nuclear inelastic absorption. Strictly speaking, an integration over infinite time interval cannot be reached in real experiments, because during a short period after synchrotron radiation flash an acquisition system is gated against strong electronic scattering. In order to analyze possible influence of this gating on energy spectra of inelastic scattering, we discuss time dependence of scattered radiation intensity.

4.2 Time dependence of scattered radiation.

Eq. (30) for intensity of scattered radiation can be presented as follows

$$I(\omega_0, t) = I_0 I_N \, |Q_{fi}(\omega_0, t)|^2 \, , \tag{46}$$

where

$$Q_{fi}(\omega_0, t) = \int \frac{d\omega}{2\pi} \exp(-i\omega t) \tilde{L}_{fi}(\mathbf{k}_1, -\mathbf{k}_0, \omega_0 - \omega_r + \omega + i\gamma) P(\omega) \, . \tag{47}$$

Substituting (28) we write this expression in the form

$$Q_{fi}(\omega_0, t) = \int_0^t dt' \exp(i[\omega_0 - \omega_r + i\gamma]t') L_{fi}(\mathbf{k}_1, -\mathbf{k}_0, t') \tilde{P}(t - t') \, . \tag{48}$$

Here we use eq.(8) and that $\tilde{P}(t) = 0$ for $t < 0$. As follows from eq. (48), the intensity vanishes at $t = 0$. Making a replacement of the integration variable $t' \to t - t'$ and using eq. (25) we obtain

$$Q_{fi}(\omega_0, t) = e^{-\gamma t} e^{i(\omega_0 - \omega_r)t} \sum_m \langle \chi_f \, | e^{-i\mathbf{k}_1 \mathbf{r}_n} | \chi_m \rangle \langle \chi_m \, | e^{i\mathbf{k}_0 \mathbf{r}_n} | \chi_i \rangle e^{-i\omega_{mi}^{(p)} t}$$

$$\times \int_0^t dt' \exp(i[\omega_r + \omega_{mi}^{(p)} - \omega_0]t') \, e^{\gamma t'} \, \tilde{P}(t') \, . \tag{49}$$

General evaluation of this expression requires an explicit form of the response function of the monochromator. We will consider the case where the energy width of the instrumental function is much larger than that of nuclear resonance. Therefore, the response function $\tilde{P}(t)$ decreases significantly on the characteristic time t_p, which is much less than the life time of excited nuclear state $t_0 = 2/\gamma$. Then the function $\exp(\gamma t')$ under the integral can be approximated by unity. It is evident that within the time interval $0 < t < t_p$ the integral in eq. (49) increases with time and it becomes constant when $t \gg t_p$. In this latter region we can replace the upper limit by ∞. Omitting also a constant phase factor which does not influence the intensity, we obtain

$$Q_{fi}(\omega_0, t) = e^{-\gamma t} \sum_m \langle \chi_f \, | e^{-i\mathbf{k}_1 \mathbf{r}_n} | \chi_m \rangle \langle \chi_m \, | e^{i\mathbf{k}_0 \mathbf{r}_n} | \chi_i \rangle e^{-i\omega_{mi}^{(p)} t} P(\omega_r + \omega_{mi}^{(p)} - \omega_0) \, . \tag{50}$$

Repeating derivation as discussed above, we arrive to the following expression for the intensity summarized over the final states and averaged over the initial states

$$\overline{I(\omega_0, t)} = I_0 \exp(-t/t_0) I_N \sum_{i,f} g_i \, |\langle \chi_f | \exp(i\mathbf{k}_0 \mathbf{r}_n)|\chi_i\rangle|^2 I_M(\omega_r + \omega_{fi}^{(p)} - \omega_0) \, . \tag{51}$$

We obtain that within the time region $t \gg t_p$ the intensity decreases in an expected way, i.e. exponentially with the life time of the excited nuclear state.

Eq. (51) is similar to eq. (39), derived above for the case of nuclear inelastic absorption. Therefore, following the same approach, we obtain

$$\overline{I(\omega_0, t)} = I_0 \exp(-t/t_0) I_N \int dt' \exp(-i[\omega_0 - \omega_r]t') F(\mathbf{k}_0, t') \tilde{I}_M(t') \, . \tag{52}$$

If the life time of excited nuclear state is, as we assume here, much longer that the duration of the flash of incident radiation provided by the monochromator, we can neglect time dependence of the function $R(t)$ in eq. (44) and replace $R(t)$ by $R(0)$. Then the integrated over time intensity (52) coincides with the approximate expression (44), obtained by other way.

Time dependence of $R(t)$ in eq.(44) allows one to study correct behaviour of the delayed radiation for very small delay time inside the flash provided by monochromator. Within this time interval the intensity of scattered radiation increases from zero to a certain maximal value. When the flash is over, the major effect is the delayed re-emission of excited nuclear state which decays exponentially with the time.

5. Effect of hyperfine structure of nuclear levels.

In this section we consider more complicated case, where nuclear levels of both ground and excited states are split by hyperfine interaction. In eqs. (19), (22) and further down we have to replace ω_i by $\omega_i + \Delta\omega_\alpha^{(i)}$, ω_j by $\omega_j + \Delta\omega_\beta^{(j)}$, ω_f by $\omega_f + \Delta\omega_\gamma^{(f)}$, where indexes α, β and γ denote various sublevels of hyperfine structure for the initial, intermediate, and final states, respectively. Accordingly, nuclear wave functions have indexes $|\phi_{i\alpha}\rangle$ and so on. Eq. (22) for the scattering matrix becomes

$$M_{fi}(\mathbf{r},t;\mathbf{r}',t')=\frac{i\exp(i\omega_{f\gamma,i\alpha}^{(n)}t)}{\hbar c}\int\frac{d\mathbf{k}d\mathbf{k}'}{(2\pi)^6}e^{i\mathbf{k}\mathbf{r}+i\mathbf{k}'\mathbf{r}'}N_{fi}^{\gamma\alpha}(\mathbf{k},\mathbf{k}',t-t')$$
$$\times\exp[i(-\omega_{ji}^{(n)}+i\gamma_j)(t-t')]e^{-i\mathbf{k}\mathbf{r}_n(t)}e^{-i\mathbf{k}'\mathbf{r}_n(t')}\,,\tag{53}$$

where $\omega_{f\gamma,i\alpha}=\omega_f+\Delta\omega_\gamma^{(f)}-\omega_i-\Delta\omega_\alpha^{(i)}$, and the nuclear function $N_{fi}^{\gamma\alpha}(\mathbf{k},\mathbf{k}',t)$ acquire the time dependence

$$N_{fi}^{\gamma\alpha}(\mathbf{k},\mathbf{k}',t)=\sum_\beta\langle\phi_{f\gamma}|\,\mathbf{e}_f\hat{\mathbf{j}}_n(\mathbf{k})|\phi_{j\beta}\rangle\langle\phi_{j\beta}|\,\mathbf{e}_i\hat{\mathbf{j}}_n(\mathbf{k}')|\phi_{i\alpha}\rangle\exp\left(i\Delta\omega_{j\beta,i\alpha}t\right)\,,\tag{54}$$

where $\Delta\omega_{j\beta,i\alpha}=\Delta\omega_\beta^{(j)}-\Delta\omega_\alpha^{(i)}$.

Repeating the derivation discussed above, it is easy to obtain the general expression for the time dependence of intensity as

$$I(\omega_0,t)=I_0\sum_{\alpha,\gamma}g_\alpha\left|\tilde{Q}_{fi}^{\gamma\alpha}(\omega_0,t)\right|^2\,,\tag{55}$$

where g_α is the statistical weight of the state $(i\alpha)$, and

$$\tilde{Q}_{fi}^{\gamma\alpha}(\omega_0,t)=\int_0^t dt'\exp(i[\omega_0-\omega_r+i\gamma]t')\,N_{fi}^{\gamma\alpha}(\mathbf{k}_1,-\mathbf{k}_0,t')$$
$$\times L_{fi}(\mathbf{k}_1,-\mathbf{k}_0,t')\tilde{P}(t-t')\,.\tag{56}$$

We note that hyperfine splitting can significantly exceed the width of the nuclear resonance. Nevertheless, the characteristic time $t_N\approx 1/\Delta\omega$ of nuclear function

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variation is much larger than the characteristic time t_p of the response function of the monochromator. Therefore at $t \gg t_p$ we can replace the lower limit of integration by $-\infty$ and the function, $\exp(-\gamma t') N_{fi}^{\gamma\alpha}(\mathbf{k}_1, -\mathbf{k}_0, t')$ by its value at the upper limit t.

As a result, instead of (52), we obtain the generalized equation

$$\overline{I(\omega_0, t)} = I_0 \exp(-t/t_0) I_N(t) \int dt' \exp(-i[\omega_0 - \omega_r]t') F(\mathbf{k}_0, t') \tilde{I}_M(t') \,, \qquad (57)$$

where

$$I_N(t) = \sum_{\alpha, \gamma} g_\alpha \left| N_{fi}^{\gamma\alpha}(\mathbf{k}_1, -\mathbf{k}_0, t) \right|^2 . \qquad (58)$$

To obtain the intensity $I_N(t)$ integrated over the angle of exit of scattered photons, it is necessary to integrate over directions of the vector $\mathbf{k}_1$. The function $I_N(t)$ does not depend on the resonance condition, in particular, on the value $(\omega_0 - \omega_r)$. Therefore it does not influence the energy spectrum of incoherent nuclear inelastic scattering.

6. Conclusion

We present the theoretical analysis of energy and time dependence of incoherent nuclear inelastic resonant scattering of synchrotron radiation (SR) accompanied by absorption and emission of phonons in crystal lattice. The theory is based on the Maxwell's equations and the time dependent perturbation theory of the quantum mechanics. This allows us to analyze explicitly physical nature of the considered processes.

In order to treat the case of incoherent scattering, we assume that, though the scattering process is coherent in general, the conditions of measurements do not allow experimental observation of the interference term in the intensity of scattered radiation due to its smearing out during averaging over various parameters. This concept of incoherent scattering significantly simplify calculations, because it avoids calculations of the interference term which would in any event vanish during averaging. Our results show that if measurements are performed without energy or angular analysis of scattered radiation, the dependence of its intensity on the energy of incident radiation is identical to that of nuclear inelastic absorption. Formally, this is caused by averaging of scattering intensity over final states of phonon system.

The measured spectra are influenced only by processes of absorption and emission of phonons only during the nuclear excitation by the SR flash. We assume that x rays emitted by various electrons in a storage ring are incoherent, and radiation from single electron can be considered as instantaneous. This assumption is based on the experimental conditions, where the intensity of scattered radiation is accumulated after many SR flashes, whereas only one photon is recorded after each SR flash. We show that the possibility to measure the density of phonon states under these conditions arises from the property of monochromator to delay the initially instantaneous SR flash up to the time interval which significantly exceeds the period of nuclear vibrations. It is sufficient to describe the case of incoherent scattering within the

first Born approximation. Then the calculation is reduced to thermal averaging of the phase factor depending on the nuclear coordinates at the instant of photon both absorption and emission. Within this approach, the time correlation function can be calculated with a proper account for multi-phonon processes, whereas treating the multi-phonon processes in the case of coherent scattering is significantly more complicated problem.

<p style="text-align:center">*   *   *</p>

References

1. R. L. Mössbauer, Z. Physik, **151**, 124 (1958)
2. Yu. Kagan, Hyp. Inter., **123/124**, 83 (1999)
3. J. P. Hannon, G. T. Trammell, Hyp. Inter., **123/124**, 127 (1999)
4. W. M. Visscher, Ann. Phys., **9**, 194 (1960)
5. K. S. Singwi, A. Sjölander, Phys. Rev., **120**, 1093 (1960)
6. Yu. M. Kagan, Yu. A. Iosilevskii, Zhurn. Exp. Teor. Fiz., **42**, 259 (1962); **44**, 284 (1963) (in russian)
7. A. I. Chumakov, W. Sturhahn, Hyp. Inter., **123/124**, 781 (1999)
8. V. G. Kohn, A. I. Chumakov, R. Rüffer, Phys. Rev. B, **58**, 8437 (1998)
9. W. Sturhahn, V. G. Kohn, Hyp. Inter., **123/124**, 369 (1999)
10. M. v. Laue, *Röntgenstrahlinterferenzen*, 1941; 1960, Frankfurt.
11. W. Heitler, *The quantum theory of radiation*, 1944, Oxford University Press, N.Y.
12. J. J. Sakurai, *Advanced quantum mechanics*, 1967, Addison-Wesley, USA.
13. U. v. Bürck, Hyp. Inter., **123/124**, 483 (1999)
14. V. Kohn, I. Snigireva, A. Snigirev, Phys. Rev. Lett., **85**, 2745 (2000)
15. I. M. Ternov, Usp. Fiz. Nauk, **165**, 429 (1995) (in russian)
16. G. Margaritondo, J. Synchrotron Rad., **2**, 148 (1995)
17. A. I. Chumakov, A. Barla, R. Rüffer, J. Metge, H. F. Grünsteudel, H. Grünsteudel, J. Plessel, H. Winkelmann, M. M. Abd-Elmeguid, Phys. Rev. B, **58**, 254 (1998)
18. V. G. Kohn, A. I. Chumakov, Hyp. Inter., **125**, 205 (2000)